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USE OF ULTRACOUSTIC DATA FOR THE CALCULATION OF THE THERMAL CONDUCTIVITY OF GLASSY SUBSTANCES AT HIGH TEMPERATURES

by

M. V. Bessonov

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13. ABSTRACT

The determination of the true values of the thermal conductivity of semitransparent glassy substances at high temperatures is rendered difficult by the influence of radiative heat transfer. The thermal conductivity gamma of semitransparent substances can be found from acoustic data on the basis of the Debye or Bridgman formulae connecting gamma with the mean velocity of the elastic waves. An investigation of the velocity of propagation of ultrasound in glassy substances (colophony and sodium-silicate glass) with a view to determine their gamma, is described. The velocity of the thermal elastic waves given in a melt deviates substantially, due to the relaxation dispersion, from the velocity of the ultrasound. To a first approximation given can be determined by a linear extrapolation of the velocity in a solid substance for high temperatures. This method was tested on a substance with a known value of gamma (colophony) and used for the study of silicate glasses. The calculation, conducted by the Debye, Bridgman and Predvoditelev-Varhaftig formulae, does not give the considerable increase in gamma with temperature. Orig. art. has: 4 figures.

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By: M. V. Bessonov

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USE OF ULTRAACOUSTIC DATA FOR THE CALCULATION OF THERMAL CONDUCTIVITY OF GLASSY SUBSTANCES AT HIGH TEMPERATURES

[Article by M. V. Bessonov; Moscow, Primenenie Ul'traakustiki k Issledovaniyu Veshchestva, Russian, Issue 13, 1961, pp. 165-170.]

Heat transfer in highly viscous media which are somewhat transparent to heat radiation is accomplished by both conduction and radiation. The theoretical treatment of heat transfer by simultaneous conduction and radiation is exceedingly complex. Significant theoretical results have been achieved only recently. Articles are being published now which permit complete calculation of heat transfer by simultaneous conduction and radiation if the thermal and radiation transmission characteristics of the medium are known[1,2]. Among these is the coefficient of molecular thermal conductivity. However, reliable values of this quantity for semitransparent media, such as glasses, are not available. Only effective values are given, which include radiation heat transfer; these are strongly dependent on experimental conditions. In one experiment[3], an attempt was made to determine the true thermal conductivity of a certain glass at 850 -- 1450°C. The values obtained for the coefficient of thermal conductivity were two to three times larger than those at normal temperatures. The physical causes of such a significant increase in molecular thermal conductivity are uncertain, since melted silicates may be considered viscous fluids. The thermal conductivity of viscous fluids is comparatively weakly dependent on temperature, and usually decreases with increasing temperature.

It is possible to solve this physical problem and calculate the coefficient of thermal conductivity λ of semitransparent media in cases where direct measurement is very difficult. This may be accomplished with the aid of acoustic data. In a number of well known experiments attempts have been made to establish a link between thermal conductivity and other physical properties of matter. This problem was solved most successfully by Debye[4]. He conceived of heat conduction as a process of heat transfer by means of elastic waves with a frequency of 10^{12}-- 10^{13} Hz, which propagated from warmer to cooler sites. Thermal elastic waves are multiply scattered when they encounter the temperature anomalies and other physical discontinuities which characterize the mean free path l. Thus the speed of heat conduction is far slower than the waves' velocity of propagation. Debye obtained the following expression for λ:

$$\lambda = \frac{1}{4} \rho c l \bar{u} \qquad (1)$$

where ρ=density, c=heat capacity, and $\bar{u}$= mean velocity of the thermal elastic waves, defined by Debye in the following relation

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between the velocity of longitudinal waves u_e and the velocity of transverse waves u_t:

$$\frac{3}{\bar{u}} = \frac{1}{u_e} + \frac{2}{u_t}$$

Though the Debye equation is hard to apply in the case of crystals because of the complexity of determining l, for amorphous glassy substances this difficulty is lessened. For these substances l is determined from the mean distance between the molecules or molecular complexes which take part in thermal motion; it is not temperature dependent. Besides relation (1), there is also the Bridzhmen formula [5] which showed that experimental results concerning thermal conductivity of fluids are satisfactorily described by the simple relation:

$$\lambda = 3k\bar{u}\delta^{-2} \qquad (2)$$

Here k is Boltzmann's constant, δ is the distance between molecules.

Bridzhmen also gave an elementary consequence of formula (2). Equations (1) and (2) contain the velocity of sound; its temperature dependence determines the change of thermal conductivity with temperature. However, the velocity of sound in glasses is unknown at temperatures higher than 500 -- 600°. An experiment was conducted to investigate the velocity of propagation of ultrasound in glassy substances, in order to determine their thermal conductivity. Sodium silicate glass was among those tested. A description of the method and the experimental set-up is given in issue 8 of this journal [6].

The temperature dependence of the velocity of ultrasound in rosin and in silicate glass is shown in Figures 1 and 2.

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Fig. 1 The temperature dependence of the velocity of ultrasound in rosin.(6)
u_0= velocity for $\nu \to 0$,
u_∞= velocity for $\nu \to \infty$)

KEY: (1) u/msec); (2) MHz.

Fig. 2 The temperature dependence of the velocity of ultrasound in sodium-silicate glass.
u_0= velocity for $\nu \to 0$,
u_∞= velocity for $\nu \to \infty$)
KEY: (1) kcal/m-hr °C

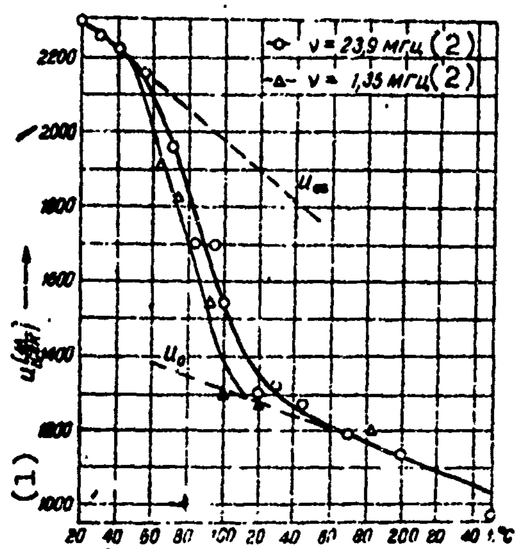

Fig. 1

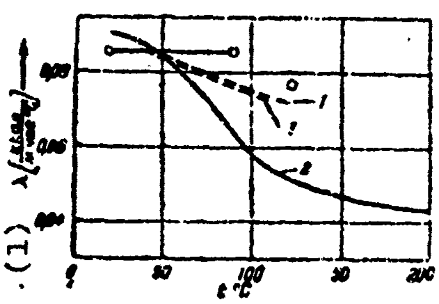

Fig. 2

The data show that when glass changes from the solid to the liquid state, a decrease in the velocity of ultrasound by a factor of 2.5 — 3 is associated with the transition. This decrease is accompanied by dispersion which we observed directly in rosin [6], and which other authors have observed during the vitrification of viscous fluids [7,8]. These phenomena are explained qualitatively by relaxation theory. Direct measurement and calculation of the thermal conductivity of rosin also indicates significant dispersion. In fact, from formula (2) and experimental values of the velocity of sound, the thermal conductivity of rosin decreases sharply at 60°C (Fig. 3)

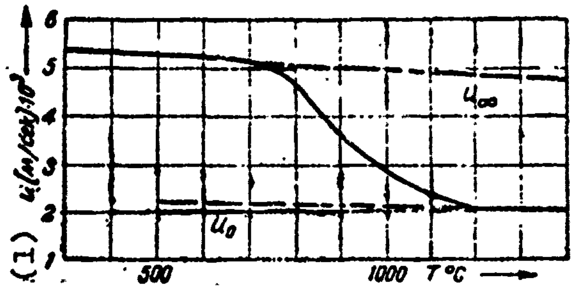

Fig. 3. The temperature dependence of the coefficient of thermal conductivity of rosin (0- experiment; 1- calculated from formula (2) and the velocity of thermal elastic waves, determined from the velocity of ultrasound in solid rosin; 2- calculated from formula (2) and the velocity of ultrasound; 3- calculated from the formula of Predvoditelev-Vargaftik $\lambda = B \cdot \beta^{+3}$, B=const)

KEY: (1) u(m/sec$\cdot 10^3$.

This is contradicted by the experimental data of E. Kuvshinskiy [9], which show that λ_0 of rosin remains constant $\pm 5\%$ in the temperature interval 20 -- 90°C. The discrepancy between calculated and experimental values must be explained by the fact that the mean velocity $\bar{u}$ is determined from the velocity of ultrasound in the melted state. However, the velocity of thermal elastic waves u_∞ as a consequence of relaxation dispersion differs significantly from the velocity of ultrasound. An estimate of the relaxation time in rosin from experimental values of the velocity of ultrasound shows that its magnitude for frequencies of 10^{12}-10^{13} Hz is much greater than unity, at least up to 120 -- 130°C. Consequently, the velocity of the thermal elastic waves remains practically the same as it is in the solid state. A first approximation of the velocity u_∞ may be determined by linear extrapolation to high temperatures of the velocity in solid rosin. For this case the value of λ calculated from formula (2) is close to the experimental value (Fig.3).

The best results from calculation may be obtained from Debye's formula (1), according to which some decrease in velocity is compensated for by an increase in heat capacity; this takes place during the softening of glasses.

The calculation of the coefficient of thermal conductivity of again thus shows that the velocity u_{∞} for glassy substances should be determined from the velocity in the solid state. This method of calculation was employed to determine the thermal conductivity of silicate glasses. The results of the calculation of the coefficient of thermal conductivity for three sodium silicate glasses, for which the velocity of sound was measured in the solid state [10], are shown in Fig. 4. These glasses are similar in composition to those tested in article [3].

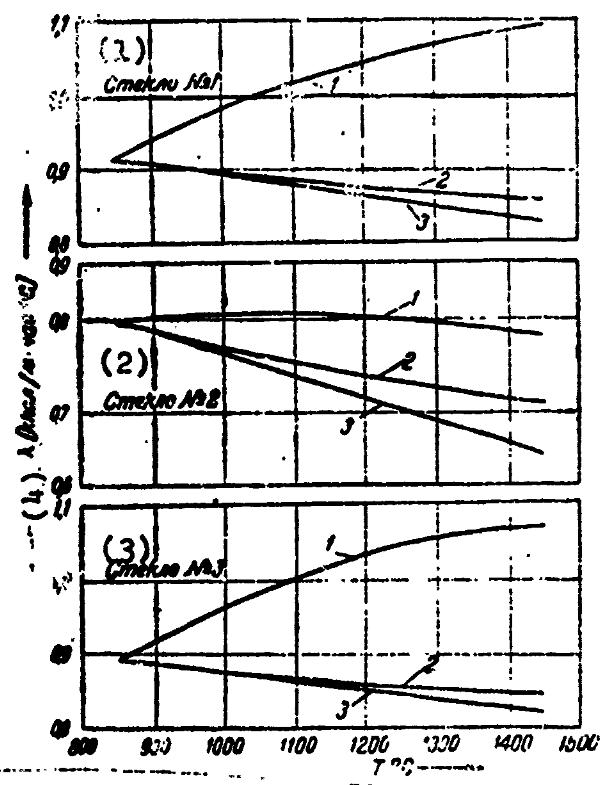

Fig. 4. Calculation of the coefficient of thermal conductivity of sodium silicate glass. (1 – by Debye formula; 2 – by formula of Predvoditelev-Vargaftik; 3 – by the Bridzhmen formula).

KEY: (1) Glass No. 1; (2) Glass No. 2; (3) Glass No. 3; (4) kcal/m-hr °C.

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The calculation of thermal conductivity was carried out here according to the formulas of Debye, Bridzhmen, and Predvoditelev-Vargaftik. Not one of the formulas indicates a significant increase in thermal conductivity when compared to the experimental data cited by Gutop [3], which show an increase in λ by a factor of 2.5 — 3 as the temperature is raised from 800 — 1450°C.

The analysis of this data in the light of new theories of heat transfer is still incomplete. There is no reason, however, to consider incorrect the values of thermal currents observed experimentally. Therefore, experimental values of thermal currents may serve as a standard for checking theoretical calculations of λ. The values of the coefficient of thermal conductivity λ determined from the velocity of sound are used with formulas[2]to give theoretical predictions of the thermal currents, which are then compared with experimental results. The results of such comparisons [11] show that best agreement is obtained if λ is calculated from the Debye fórmula. This indicates that of the formulas considered, Debye's formula gives the best estimate of the temperature dependence of the thermal conductivity of silicate glasses.

The following results are presented in conclusion:

1. The study of the propagation of ultrasound in glasses has led to the determination of the temperature dependence of the thermal conductivity of glassy substances in a viscous liquid state.

2. Data concerning the strong temperature dependence of the thermal conductivity of softened glasses have not been confirmed.

3. The possibility of applying acoustic data to the calculation of thermal conductivity has been demonstrated; with the aid of new theories of heat transfer this permits the calculation of thermal currents in semi-transparent media.

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